

Magneto-optical and Magneto-electric Effects of Topological Insulators in Quantizing Magnetic Fields

Wang-Kong Tse and A. H. MacDonald

Department of Physics, University of Texas, Austin, Texas 78712, USA

We develop a theory of the magneto-optical and magneto-electric properties of a topological insulator thin film in the presence of a quantizing external magnetic field. We find that low-frequency magneto-optical properties depend only on the sum of top and bottom surface Dirac-cone filling factors ν_T and ν_B , whereas the low-frequency magneto-electric response depends only on the difference. The Faraday rotation is quantized in integer multiples of the fine structure constant and the Kerr effect exhibits a $\pi/2$ rotation. Strongly enhanced cyclotron-resonance features appear at higher frequencies that are sensitive to the filling factors of both surfaces. When the product of the bulk conductivity and the film thickness in e^2/h units is small compared to α , magneto-optical properties are only weakly dependent on accidental doping in the interior of the film.

PACS numbers: 78.20.Ls, 73.43.-f, 75.85.+t, 78.67.-n

Introduction— Topological insulators (TIs) are a recently identified [1] new class of materials. Three-dimensional TIs have insulating bulks and metallic surfaces with an odd number of Dirac cones that are responsible for most unique TI properties. Angle-resolved photoemission spectroscopy (ARPES) experiments have established that several strongly spin-orbit coupled materials [2] exhibit TI properties. In this paper we develop a theory of the magneto-optical and magneto-electric properties of TI thin films in the presence of a perpendicular external magnetic field.

Our work is motivated in part by potential advantages of magneto-optical over transport [3] characterization in isolating TI surface properties from bulk contamination due to unintended doping. Since Landau level (LL) quantization of the TI's surface Dirac cones has recently been established by STM experiments [4], it should be possible to detect surface quantum Hall effects optically, even when parallel bulk conduction is present. In the quantum Hall regime, we find that the low-frequency Faraday effect is *quantized* in integer multiples of the fine structure constant, while the Kerr effect displays the same [5] giant $\pi/2$ rotation relative to the incident polarization direction that appears when time reversal is broken by exchange coupling. At higher frequencies, we find strong cyclotron resonance features in both Faraday and Kerr spectra.

One goal of our work is to clarify how the magneto-electric effects peculiar to TIs [5–9] are reflected in their thin film magneto-optical properties. We show that low-frequency TI magneto-optical response in the quantum Hall regime depends on the sum of top and bottom surface filling factors, whereas the magneto-electric response of film polarization to an external magnetic field depends on the filling factor difference. We argue that coupling between electric and magnetic fields in the presence of a TI material is most usefully regarded as a property of its surfaces, not of its bulk.

Low-frequency Magneto-electric and Magneto-optical Re-

sponse— The response of a TI thin film to an external magnetic field is dominated by [10] its Dirac-cone surface states. When the Dirac cone quantum Hall effect is well developed, its Hall conductivity σ_{xy} has quantized plateau values with half-odd-integer values in e^2/h units. The longitudinal resistivity vanishes on the Hall plateaus, but is non-zero on the *risers* between Hall plateaus. The risers between the $(n - 1/2)(e^2/h)$ and $(n + 1/2)(e^2/h)$ plateaus occur near integer values of the filling factor $\nu = n \in \mathbb{Z}$. The Dirac cone's quantum Hall effect has so far been cleanly observed [11] only in two-dimensional graphene, in which four separate Dirac cones conduct in parallel.

The Streda [12] formula,

$$\sigma_{xy} = ec(\partial N/\partial B) = ec(\partial M/\partial \mu), \quad (1)$$

which is valid at Hall plateau centers, implies a relationship between surface conductivities and the magneto-electric response of a TI thin film. In Eq. (1) N is the two-dimensional electron density, B the external magnetic field, M the orbital magnetization, and μ the chemical potential. We define the electric polarization P per unit volume of a TI thin film in terms of the difference between the surface charge densities accumulated on the top (T) and bottom (B) surfaces $P = e(N_T - N_B)/2$. It follows that for plateaus characterized by ν_T and ν_B the magneto-electric susceptibility

$$\chi_{ME} = 4\pi\partial P/\partial B = (\nu_T - \nu_B)\alpha, \quad (2)$$

depends only on the filling factor difference between surfaces ($\alpha = e^2/hc$ is the fine structure constant). Recent experiments have demonstrated [13] the possibility of tuning the surface carrier densities systematically by surface doping or gating. At a fixed magnetic field, the top and bottom surfaces need not be on the same Hall plateau. Provided that the bulk resistance is sufficiently large, χ_{ME} can be measured by contacting top and bottom surfaces separately and detecting voltages induced

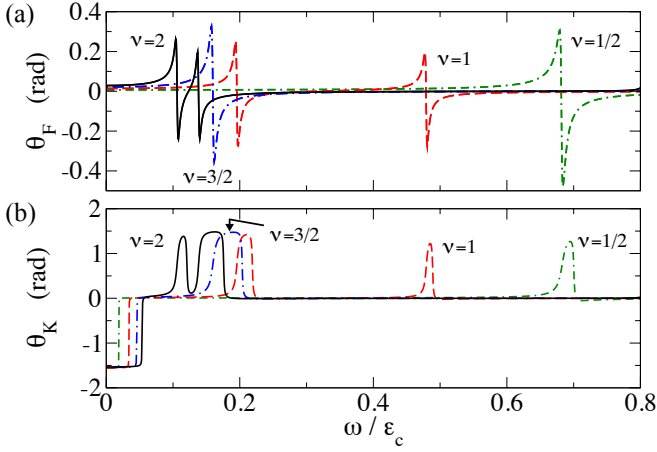


FIG. 1: (Color online). (a). Faraday rotation θ_F versus frequency ω/ε_c at equal filling factors on both surfaces $\nu = 1/2$ (green), $\nu = 1$ (red), $\nu = 3/2$ (blue), and $\nu = 2$ (black). The densities on both surfaces are $N_{T,B} = 5 \times 10^{11} \text{ cm}^{-2}$. (For this density the filling factor is given by $\nu = 20.81/B [\text{Tesla}]$.) We choose a 30 nm-thick Bi_2Se_3 film as a prime example of TIs with a large bulk band gap $E_g = 0.35 \text{ eV}$. The Fermi velocity is $v = 5 \times 10^5 \text{ cm}^{-2}$ and the dielectric constant $\varepsilon = 29$. (b). Kerr rotation θ_K versus frequency at the same filling factors.

by magnetic field variation. If sample quality can be improved sufficiently, TI thin films might be useful as magnetic field sensors.

The TI's surface quantum Hall effect can also be probed by Faraday and Kerr angle measurements. In the low frequency regime the electromagnetic wavelength is much longer than the film thickness. Faraday's and Ampère's Laws then imply that the electric field is spatially constant across the film, while the magnetic field jumps by a value proportional to the current integrated across the TI film. Allowing for a bulk conductivity Σ due to unintended doping [3] and assuming a small bulk Hall angle we find that the low-frequency quantum-Hall-regime Faraday and Kerr angles depend on the TI surface properties only through $\nu_T + \nu_B$:

$$\tan \theta_F = \frac{(\nu_T + \nu_B) \alpha}{1 + 2\pi \Sigma d/c}, \quad (3)$$

$$\tan \theta_K = \frac{4(\nu_T + \nu_B) \alpha}{1 - (1 + 4\pi \Sigma d/c)^2 - [2(\nu_T + \nu_B) \alpha]^2}. \quad (4)$$

The bulk carriers enter as an *effective* longitudinal surface conductivity Σd , where d is the film thickness. When the bulk conductivity is sufficiently small so that $\Sigma d/(e^2/h) \lesssim \alpha$, the Kerr angle exhibits a universal full-quarter rotation $\theta_K \simeq \pm \pi/2$. When $\Sigma d/(e^2/h) \ll 1/\alpha$, the Faraday angle is quantized in integer multiples of the fine structure constant

$$\theta_F \simeq (\nu_T + \nu_B) \alpha. \quad (5)$$

Given the rapid progress in TI film quality, these regimes should be within reach experimentally; in particular,

the less stringent condition for Faraday angle quantization should be currently accessible. Since the magneto-electric polarizability Eq. (2) yields the filling factor difference, and the Faraday angle Eq. (5) yields the filling factor sum, measurement of both quantities could allow the filling factors $\nu_{T,B}$ to be extracted individually.

Dirac-Cone ac Conductivity— Outside of the long-wavelength limit, TI thin-film magneto-optical properties depend on the finite-frequency Dirac-cone conductivity which we now evaluate microscopically. The high-frequency signal consists of resonances at inter-LL transition frequencies. We neglect optical phonon contributions to the conductivity which are not expected to be significantly dependent on magnetic field strength. In an external magnetic field the Dirac-cone Hamiltonians for the top (T) and bottom (B) surfaces are $H = (-1)^L [v\boldsymbol{\tau} \cdot (-i\nabla + e\mathbf{A}/c) + V/2] + \Delta\tau_z$, where $\boldsymbol{\tau}$ is the spin Pauli matrix vector, $\mathbf{A} = (0, Bx)$ is the vector potential, $\Delta = g\mu_B B/2$ is the Zeeman coupling, V accounts for a possible potential difference between top and bottom surfaces due to doping or external gates, and $L = 0, 1$ for the top (0) and bottom (1) surfaces. The LLs are labeled by integers n and for $n \neq 0$ have eigenenergies (relative to the Dirac point energies $(-1)^L V/2$) $\varepsilon_n = \text{sgn}(n) \sqrt{2v^2|n|/\ell_B^2 + \Delta^2}$, where $\ell_B = \sqrt{c/e|B|}$ is the magnetic length. In the $n = 0$ LL spins are aligned with the perpendicular field and $\varepsilon_0 = -\Delta$. For convenience we rewrite the LL index as $n = sm$, where $m = 0, 1, 2, \dots, N_c$ and $s = \pm 1$ for electron-like and hole-like LLs. $N_c \simeq \ell_B^2(\varepsilon_c^2 - \Delta^2)/2v^2$ is the largest LL index with an energy smaller than the ultraviolet cut-off ε_c . We choose $\varepsilon_c = E_g/2$ where E_g is the bulk band gap.

Using the Kubo formalism we find that in the quantum Hall regime ($\Omega_B \tau \gg 1$, where $1/\tau$ the quasiparticle lifetime broadening and $\Omega_B = v/\ell_B$ is a characteristic frequency typical of the LLs spacing) the conductivity in $e^2/h = \alpha c$ units is given by

$$\sigma_{\alpha\beta}(\omega) = \quad (6)$$

$$\frac{v^2}{2\pi\ell_B^2} \text{sgn}(B) \sum_{m=0}^{N_c-1} \sum_{s,s'=\pm 1} \frac{f_{sm} - f_{s'(m+1)}}{\varepsilon_{sm} - \varepsilon_{s'(m+1)}} \Gamma_{\alpha\beta}^{s,s'}(m, \omega),$$

Here $\alpha, \beta = \{x, y\}$, f_{sm} is a Fermi factor, and

$$\Gamma_{\{xx\}}^{s,s'}(m, \omega) = - \begin{Bmatrix} i \\ 1 \end{Bmatrix} \mathcal{C}_{\uparrow s'(m+1)}^2 \mathcal{C}_{\downarrow sm}^2 \left(\frac{1}{\omega - \varepsilon_{sm} + \varepsilon_{s'(m+1)} + i/2\tau} \pm \frac{1}{\omega + \varepsilon_{sm} - \varepsilon_{s'(m+1)} + i/2\tau} \right). \quad (7)$$

In Eq. (7), the LL eigenspinors are $\mathcal{C}_{\uparrow 0} = 0$, $\mathcal{C}_{\downarrow 0} = 1/\sqrt{2}$, and for $m \neq 0$ $\mathcal{C}_{\uparrow sm} = s\sqrt{\varepsilon_m} + s\Delta/\sqrt{2\varepsilon_m}$, and $\mathcal{C}_{\downarrow sm} = \sqrt{\varepsilon_m} - s\Delta/\sqrt{2\varepsilon_m}$. Eqs. (6)-(7) express $\sigma_{\alpha\beta}$ as a sum over interband and intraband dipole-allowed transitions which satisfy $|n'| - |n| = \pm 1$. In the $\omega = 0$ limit Eq. (6) yields correct half-quantized plateau values for the Hall conductivity.

TI Thin Film Cyclotron Resonance—Resonances at transition energies are responsible for dips and peaks in transmission and reflection spectra respectively, and give rise to associated characteristic features in Faraday and Kerr spectra. Fig. 1a shows the frequency dependence of the Faraday rotation for several different filling factors. These results are obtained by combining the *ac* Dirac-cone conductivity with the scattering matrix formalism described in Ref. [5]. We find that θ_F exhibits sharp cyclotron resonance peaks and changes sign near each allowed transition frequency. At half-odd-integer filling factors there is a single dipole-allowed intraband transition; at other filling factors there are two allowed resonances at different frequencies associated with transitions into and out of the partially filled LL. This behavior is completely different from that of an ordinary two-dimensional electron system where all dipole-allowed transitions have the same energy. Interband transitions from hole-like to electron-like LLs are weaker and appear in the same energy range as the intraband transitions only if the field is weak and the filling factor is small.

The Kerr angle θ_K also shows dramatic enhancement at the cyclotron resonance frequencies (Fig. 1b). Unlike the low-frequency giant Kerr effect, the resonant peaks which correspond to absorptive LL transitions are non-universal. We note that Zeeman coupling shifts the 0th LL away from the Dirac point and therefore breaks the electron-hole (e-h) symmetry of the LL spectrum. This implies that the $n = -1 \rightarrow 0$ and $0 \rightarrow 1$ transition frequencies, $\omega_{0 \rightarrow 1} = \sqrt{2v^2/l_B^2 + \Delta^2} + \Delta$ and $\omega_{-1 \rightarrow 0} = \sqrt{2v^2/l_B^2 + \Delta^2} - \Delta$, become different. In general, θ_K is an odd function of filling factor ν if e-h symmetry is intact. When the surface filling factors $\nu_T = -\nu_B = 1/2$ are opposite, e-h asymmetry implies that cyclotron resonance peaks of opposite sign appear in θ_K at the two transition frequencies. The Kerr effect thus provides a smoking gun to detect the absence of e-h symmetry.

In Fig. 2a we plot the Kerr angle θ_K as a function of frequency and magnetic field for fixed surface carrier densities. As in the exchange coupling case [5], the giant Kerr effect survives up to a relatively large frequency which we refer to as the Kerr frequency:

$$\omega_K = \frac{2\pi\sigma_{xy}^R(0)/c}{[(\epsilon - \mu)d + (\epsilon_s - \mu_s)d_s] - 2\pi\sigma_{xx}^I(0)/c}. \quad (8)$$

Here σ_{xx}, σ_{xy} are the total surface conductivities, ϵ, μ, d are the dielectric constant, permeability ($\simeq 1$), and thickness of the TI film, ϵ_s, μ_s, d_s are the corresponding substrate values, and $'$ in σ_{xx}^I denotes a frequency derivative. It follows from Eq. (8) that ω_K is inversely proportional to the optical thickness of the TI film and the substrate. Fig. 2b illustrates the effect of varying B on ω_K . There exists an optimal value of B for which ω_K is in the terahertz range. High ω_K values are most readily achieved on ‘low- κ ’ substrate materials like SiO₂ or using free-

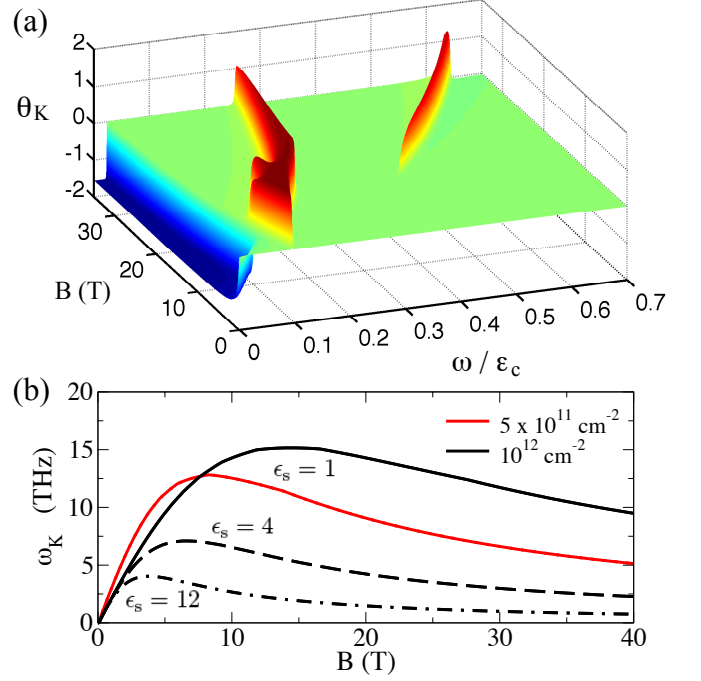


FIG. 2: (Color online) (a). Kerr angle θ_K as a function of frequency ω/ϵ_c and magnetic field B for surface densities $N_{T,B} = 5 \times 10^{11} \text{ cm}^{-2}$. Cyclotron resonance features corresponding to particular transitions are allowed when the initial Landau level is at least partially occupied and the final Landau level is at least partially empty. In the weak-field semiclassical transport regime, $\sigma_{xy} \propto B$ and θ_K vanishes as $B \rightarrow 0$. (b) Kerr frequency ω_K (see text) as a function of magnetic field for $N_{T,B} = 10^{12} \text{ cm}^{-2}$ (dark/black) for free-standing 30 nm-thick Bi₂Se₃ ($\epsilon_s = 1$), with SiO₂ substrate ($\epsilon_s = 4$), and Si substrate ($\epsilon_s = 12$). Grey/red line shows the free-standing case with $5 \times 10^{11} \text{ cm}^{-2}$. The substrate thickness $d_s = 1 \mu\text{m}$.

standing films suitable for optical studies [14]. ω_K can also be increased by surface doping.

Discussion—TIs have interesting magneto-electric and magneto-optical properties when time reversal symmetry (TRS) is broken to open up a gap in its Dirac-cone surface states. TRS can in principle be broken by exchange coupling to an insulating ferromagnet, although it is not yet established that a sufficiently strong coupling can be achieved in practice. The circumstance discussed here in which a perpendicular magnetic field is applied to a TI thin film provides an experimentally simpler and phenomenologically richer method for producing TI thin films with weak TRS breaking. The special case in which the filling factor $\nu = 1/2$ on both surfaces yields the same surface Hall conductivities as in the exchange-coupled case and therefore the same [5] low-frequency Kerr and Faraday angle results. In the magnetic field case, however, strong magneto-optical and magneto-electric effects occur over a broad range of separately controllable carrier densities on both surfaces. The top and bottom surface

Dirac cone filling factors, ν_T and ν_B , can be identified [15] from the ratios of magneto-optical resonance frequencies and from the patterns they produce in Faraday or Kerr spectra. Bulk conduction has a quantitative influence on Faraday and Kerr spectra only when $\Sigma d/(e^2/h)$, the bulk contribution to the dimensionless effective surface conductivity, is larger than about $1/\alpha$ and α respectively. In the quantum Hall regime, the Kerr angle is large whenever the filling factors sum to a non-zero value and are away from one of the integer values at which longitudinal resistance peaks occur. On quantum Hall plateaus magneto-optical properties depend only on the sum of individual surface filling factors, whereas the magneto-electric response of film polarization to field depends only on the difference.

The influence of a bulk TI material on electromagnetic fields can be represented [6] by introducing an $\mathbf{E} \cdot \mathbf{B}$ term with coefficient α_{ME} in the electromagnetic Lagrangian. The microscopic Poisson and Ampère equations then take [16, 17] the form

$$\begin{aligned}\nabla \cdot \mathbf{E} &= 4\pi\rho - \nabla\alpha_{ME} \cdot \mathbf{B}, \\ \nabla \times \mathbf{B} - (1/c)\partial\mathbf{E}/\partial t &= (4\pi/c)\mathbf{J} + \nabla\alpha_{ME} \times \mathbf{E}.\end{aligned}\quad (9)$$

When both surfaces are on Hall plateaus, Eqs. (9) with ρ and \mathbf{J} set to zero is a valid coarse-grained description of the interface provided that we set

$$d\alpha_{ME}/dz = (4\pi/c)\sigma_{xy}\delta(z) = 2\alpha\nu\delta(z). \quad (10)$$

Because [1, 8] ν can change by an integer without altering the bulk, only α_{ME} modulo 2α characterizes the bulk material. (When the bulk α_{ME} is mapped to the interval $[0, 2\alpha]$, only $\alpha_{ME} = \alpha$ is consistent with time-reversal invariance.) In the thin film geometry normally employed in experiment, however, a bulk value of α_{ME} modulo 2α is not sufficient to predict the result of a magneto-optical or magneto-electric measurement [18] because it does not specify the Hall plateau index. Moreover, in real samples with non-zero disorder the Dirac-cone surface Hall conductivities will [19] be strongly suppressed when the external magnetic field is weak. The values of α_{ME} required to correctly reproduce the low-frequency magneto-optical signal will therefore become small and vanish with the external magnetic field. In the stronger-field quantum Hall regime, the appropriate magneto-electric constant for vacuum, TI thin-film, and substrate regions can be obtained by integrating Eq. (10) to obtain $\alpha_{ME} = 0$ in the upper vacuum, $\alpha_{ME} = 2\alpha\nu_T$ in the TI thin film, and $\alpha_{ME} = 2\alpha(\nu_T + \nu_B)$ in the substrate and lower vacuum. In both cases the appropriate values of α_{ME} depend critically on surface properties. Although $\alpha_{ME} \bmod(2\alpha)$ is indeed a bulk property of a disorder-free TI, its value does not normally provide enough information to predict either magneto-optical properties or magneto-electric response.

This work was supported by the Welch grant F1473 and by DOE grant DE-FG03-02ER45985. The authors acknowledge helpful discussions with Kenneth Burch, Dennis Drew, Zhong Fang, David Vanderbilt and Shou-Cheng Zhang.

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